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Exact mode volume and Purcell factor of open optical systems

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The Purcell factor quantifies the change of the radiative decay of a dipole in an electromagnetic environment relative to free space. Designing this factor is at the heart of photonics technology, striving to develop ever smaller or less lossy optical resonators. The Purcell factor can be expressed using the electromagnetic eigenmodes of the resonators, introducing the notion of a mode volume for each mode. This approach allows to use an analytic treatment, consisting only of sums over eigenmode resonances, a so-called spectral representation. We show in the present work that the expressions for the mode volumes known and used in literature are only approximately valid for modes of high quality factor, while in general they are incorrect. We rectify this issue, introducing the exact normalization of modes. We present an analytic theory of the Purcell effect based on the exact mode normalization and resulting effective mode volume. We use a homogeneous dielectric sphere in vacuum, which is analytically solvable, to exemplify these findings.

In his short communication [1] published in 1946, E. M. Purcell introduced a factor of enhancement of the spontaneous emission rate of a dipole of frequency ω resonantly coupled to a mode in an optical resonator, which is now known as the Purcell factor (PF). He estimated this factor as

$$F = \frac{6\pi c^3 Q_n}{\omega^3 V_n}, \tag{1}$$

with the speed of light c, the quality factor Q_n of the optical mode n, and its effective volume V_n, the latter being evaluated as simply the volume of the resonator. This rough estimate of V_n has subsequently been refined [2, 3] to

$$\frac{1}{V_n} = \left[\mathbf{e}\cdot\mathbf{E}_n(\mathbf{r}_d)\right]^2, \tag{2}$$

where $\mathbf{r}_d$ is the position of the dipole and $\mathbf{e}$ the unit vector of its polarization. In this expression, the electric field of the mode $\mathbf{E}_n(\mathbf{r})$ is normalized [2] as

$$1 = \int_{\mathcal{V}} \varepsilon(\mathbf{r})\mathbf{E}_n^2(\mathbf{r})d\mathbf{r}, \tag{3}$$

where $\varepsilon(\mathbf{r})$ is the permittivity of the resonator. The integration is performed over the "quantization volume" $\mathcal{V}$. However, for an open system this volume is not defined, and simply extending $\mathcal{V}$ over the entire space leads to a diverging normalization integral since eigenmodes of an open system grow exponentially outside of the system due to their leakage. This issue was mostly ignored in the literature and patched by phenomenologically choosing a finite integration volume. Such an approach can result in relatively small errors when dealing with modes of high Q_n, as we will see later. However, the fundamental problem of calculating the exact mode normalization and thus of the mode volume remained.

Recently, a solution to this problem has been suggested. Kristensen *et al.* [4, 5] have used the normalization which was introduced by Leung *et al.* [6] for one-dimensional (1D) optical systems and later applied [7] to three dimensions. In this approach, the volume integral in Eq. (3) is complemented by a surface term and the limit of infinite volume $\mathcal{V}$ is taken:

$$1 = \lim_{\mathcal{V}\to\infty} \int_{\mathcal{V}} \varepsilon(\mathbf{r})\mathbf{E}_n^2(\mathbf{r})d\mathbf{r} + \frac{ic}{2\omega_n}\oint_{S_\mathcal{V}} \mathbf{E}_n^2(\mathbf{r})dS, \tag{4}$$

where ω_n is the mode eigenfrequency and $S_\mathcal{V}$ is the boundary of $\mathcal{V}$. It was numerically found [4] that the surface term was leading to a stable value of the integral for the rather small volumes available in 2D finite difference in time domain (FDTD) calculations. However, it was noted that this was not the case for low-Q modes, which was attributed to numerical issues. We show later that Eq. (4) is actually diverging in the limit $\mathcal{V}\to\infty$, and therefore the generalization in [7] and the normalization Eq. (4) are incorrect. Therefore, while being a cornerstone of the theory of open systems and, in particular, of the electromagnetic theory, a correct normalization of modes for determining the mode volume in Eq. (2) and thus the PF was not available in the literature.

The normalization of eigenstates is at the heart of any perturbation theory, and its absence for open systems explains also the historical fact that an exact perturbation theory was unavailable in electromagnetics until recently. Only late in 2010 such a theory called resonant-state expansion (RSE) was formulated [8] and subsequently applied to 1D, 2D and 3D systems [8–12], demonstrating its ability to accurately and efficiently calculate resonant states (RSs) – the eigenmodes – of a perturbed open optical system using the spectrum of RSs of a simpler, unperturbed one. The normalization of RSs introduced in [8] is a key element of the RSE. It paves the way for an exact calculation of mode volume and PF in optical resonators.

Here, we present a rigorous theory of the Purcell effect, based on a general exact formula for the mode volume in arbitrary optical systems, and illustrate it on the exactly solvable model of a dielectric spherical resonator.

In the weak coupling regime, the spontaneous emission rate of a quantum dipole, which is determining the local density of states and the spectral function of the resonator, has the following form [13–15], as detailed in

the supplementary material (SM):

$$\gamma(\omega) = -\frac{\omega^2}{\varepsilon_0 \hbar c^2} \boldsymbol{\mu} \cdot \mathrm{Im}\, \hat{\mathbf{G}}(\mathbf{r}_d, \mathbf{r}_d; \omega) \boldsymbol{\mu}\,, \tag{5}$$

where $\boldsymbol{\mu} = \mu \mathbf{e}$ is the electric dipole moment and ε_0 is the vacuum permittivity. The dyadic Green's function (GF) $\hat{\mathbf{G}}$ which contributes to Eq. (5) respects the outgoing wave boundary conditions and satisfies Maxwell's wave equation with a delta function source term,

$$\left(\frac{\omega^2}{c^2} \hat{\boldsymbol{\epsilon}}(\mathbf{r}) - \nabla \times \nabla \times \right) \hat{\mathbf{G}}(\mathbf{r}, \mathbf{r}'; \omega) = \hat{\mathbf{1}} \delta(\mathbf{r} - \mathbf{r}')\,, \tag{6}$$

where $\hat{\boldsymbol{\epsilon}}(\mathbf{r})$ is the dielectric tensor of the open optical system and $\hat{\mathbf{1}}$ is the unit tensor. The permeability is assumed to be $\hat{\boldsymbol{\mu}}(\mathbf{r}) = \hat{\mathbf{1}}$ throughout this paper. With modern electromagnetic software, Eq. (6) can be solved numerically by replacing the δ-like source term with a finite-size dipole. The mode volume can then be evaluated by calculating numerically the residues of the GF at its poles, as has been recently shown [16]. Such a fully numerical approach circumvents the definition of the mode volume in terms of the mode field. In an alternative method introduced by C. Sauvan *et al.* [17, 18] the mode volume is determined from the mode field calculated numerically including an artificial perfectly matched layer (PML), which is widely used in electromagnetic software packages. A PML is an absorbing layer which allows to efficiently simulate outgoing boundary conditions within a finite simulation volume. The divergence of the normalization Eq. (3) is avoided by converting the radiative losses to the outside region into absorptive losses within the simulation volume. For the example provided in Ref. [17] of a mode in a 100 nm diameter gold sphere we found good agreement of the numerical value of the mode volume with one obtained by the exact normalization method introduced in the present work, as detailed in Table S1 of the SM.

The purpose of this Letter is to rectify and complete the Purcell theory by providing the exact general formulas for the mode normalization, mode volume, and resulting PF. The normalization deals with only the mode field in a finite volume and its frequency, so that modes calculated by any available means can be used.

Inside the optical system, i.e. within the area of inhomogeneity of $\hat{\boldsymbol{\epsilon}}(\mathbf{r})$, the GF has the following spectral representation [8, 10, 12]

$$\hat{\mathbf{G}}(\mathbf{r}, \mathbf{r}'; \omega) = c^2 \sum_n \frac{\mathbf{E}_n(\mathbf{r}) \otimes \mathbf{E}_n(\mathbf{r}')}{2\omega_n(\omega - \omega_n)}\,, \tag{7}$$

in which the sum is taken over all RSs. These are the optical modes of the system, the eigen-solutions of Maxwell's wave equation satisfying the outgoing wave boundary conditions. The eigenfrequency $\omega_n = \Omega_n - i\Gamma_n$ of the RS is generally complex and contains the position Ω_n of the resonance and its half width at half maximum Γ_n. The quality factor of each RS is given by $Q_n = \Omega_n / 2\Gamma_n$. The spectral representation Eq. (7) requires that the RSs (with $\omega_n \neq 0$) are normalized according to

$$\begin{aligned} 1 \;=\; & \int_{\mathcal{V}} \mathbf{E}_n(\mathbf{r}) \cdot \hat{\boldsymbol{\epsilon}}(\mathbf{r}) \mathbf{E}_n(\mathbf{r})\, d\mathbf{r} \\ & + \frac{c^2}{2\omega_n^2} \oint_{S_{\mathcal{V}}} \left(\mathbf{E}_n \cdot \frac{\partial}{\partial s} r \frac{\partial \mathbf{E}_n}{\partial r} - r \frac{\partial \mathbf{E}_n}{\partial r} \cdot \frac{\partial \mathbf{E}_n}{\partial s} \right) dS \end{aligned} \tag{8}$$

where the first integral is taken over an arbitrary simply connected volume $\mathcal{V}$ enclosing the inhomogeneity of the system and the center of the spherical coordinates used, and the second integral is taken over its surface $S_{\mathcal{V}}$. Equation (8) is the *correct mode normalization* compatible with the spectral representation of the GF. The partial derivative $\partial f / \partial s$ is the gradient of the function $f(\mathbf{r})$ normal to the surface and $\partial f / \partial r$ is its radial derivative. The volume $\mathcal{V}$ can be arbitrarily big – both integrals in Eq. (8) grow exponentially with $\mathcal{V}$ but exactly compensate each other making the result independent of $\mathcal{V}$. Choosing $\mathcal{V}$ in the form of a sphere in 3D or a cylinder in 2D yields $\partial/\partial s = \partial/\partial r$ and a simpler form of the normalization [8, 10, 12]. A proof of the normalization Eq. (8) of RSs using a spherical volume $\mathcal{V}$ is given in Ref. [12]. Since a convenient normalization volume $\mathcal{V}$ can be different from a sphere, we have generalized here the normalization to an arbitrarily shaped simply connected volume and provided the related proof of Eq. (8) in the SM. In presence of a frequency dispersion of the permittivity, which is important e.g. in metallic resonators, the dielectric constant $\hat{\boldsymbol{\epsilon}}(\mathbf{r})$ in Eq. (8) is replaced by $\partial\left(\omega^2 \hat{\boldsymbol{\epsilon}}(\mathbf{r}, \omega)\right) / \partial\left(\omega^2\right)$, as follows from the derivation of Eq. (8) provided in the SM.

The electric field of a RS normalized by Eq. (8) then determines via Eq. (2) its *exact mode volume*. The spectral representation Eq. (7) in turn determines the PF taking into account the contribution of all significant modes. Indeed, using Eqs. (5) and (7), the exact PF in the weak coupling regime is obtained as

$$F(\omega) = \frac{\gamma(\omega)}{\gamma_0(\omega)} = \frac{3\pi c^3}{\omega} \sum_n \mathrm{Im}\, \frac{1}{V_n \omega_n (\omega_n - \omega)}\,, \tag{9}$$

where $\gamma_0 = \omega^3 \mu^2 / (6\pi \varepsilon_0 \hbar c^3)$ is the radiative decay rate of the dipole in free space [1], which can be deduced from Eq. (5) using the GF of empty space [19], as demonstrated in the SM. If a single mode n dominates in Eq. (9), the PF on resonance ($\omega = \mathrm{Re}\, \omega_n$) can be approximated as $F(\omega) \approx 6\pi c^3 Q_n / (\omega^2 \mathrm{Re}(\omega_n V_n))$. For a high-Q mode, the eigenfrequency and mode volume are approximately real, and the latter formula reproduces Purcell's result Eq. (1) when using the correct mode volume V_n.

For illustration, we have calculated the mode volume and PF of a dielectric spherical resonator with homogeneous permittivity ($\varepsilon = 4$) surrounded by vacuum, for a point dipole placed at $|\mathbf{r}_d| = 0.9a$ with direction $\mathbf{e} = (0, 0, 1)$ in spherical coordinates (see sketch in Fig. 1a). The inverse mode volume of several eigen-

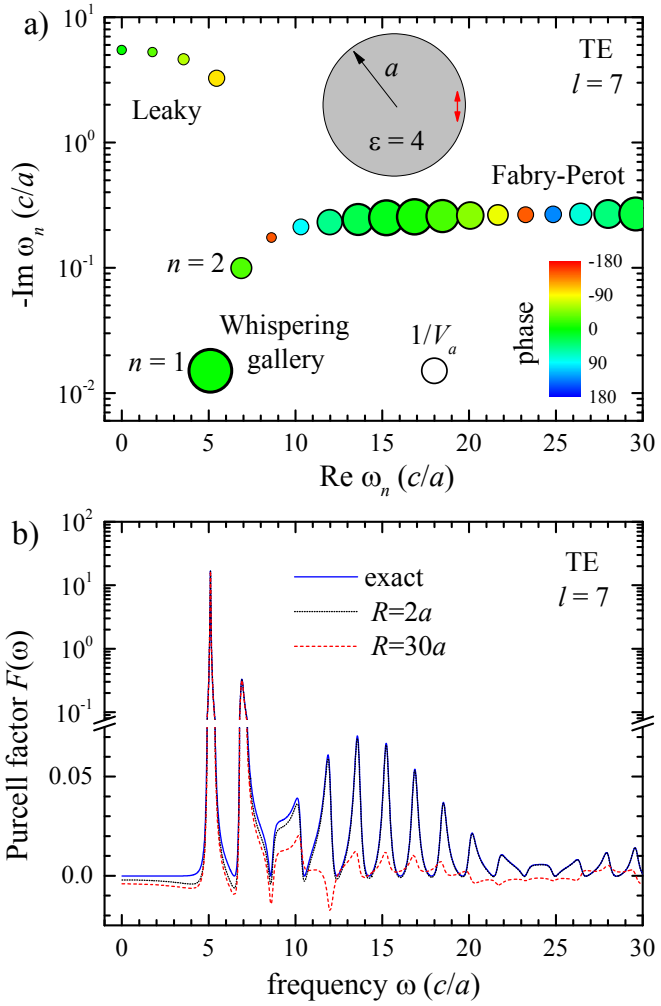

FIG. 1: (color online) a) Mode volumes for a dielectric sphere in vacuum, with permittivity $\varepsilon = 4$ and radius a, for $l = 7$ TE modes and a point dipole placed at $|\mathbf{r}_d| = 0.9a$ with direction $\mathbf{e} = (0, 0, 1)$ in spherical coordinates (see sketch). The mode volume is presented as the sum of the inverse mode volume over all degenerate states $m = -l, \ldots, l$. Its amplitude is shown by the circle area and its phase by the color. The volume of the sphere $V_a = 4\pi a^3/3$ is shown for comparison. The position of the circles in the complex frequency plane is given by the mode eigenfrequency ω_n. b) Purcell factor calculated for the geometry of a) using the exact mode normalization (blue line), and the normalization Eq. (4) evaluated for integration volumes given by a sphere of radius $R = 2a$ and $R = 30a$, as labeled.

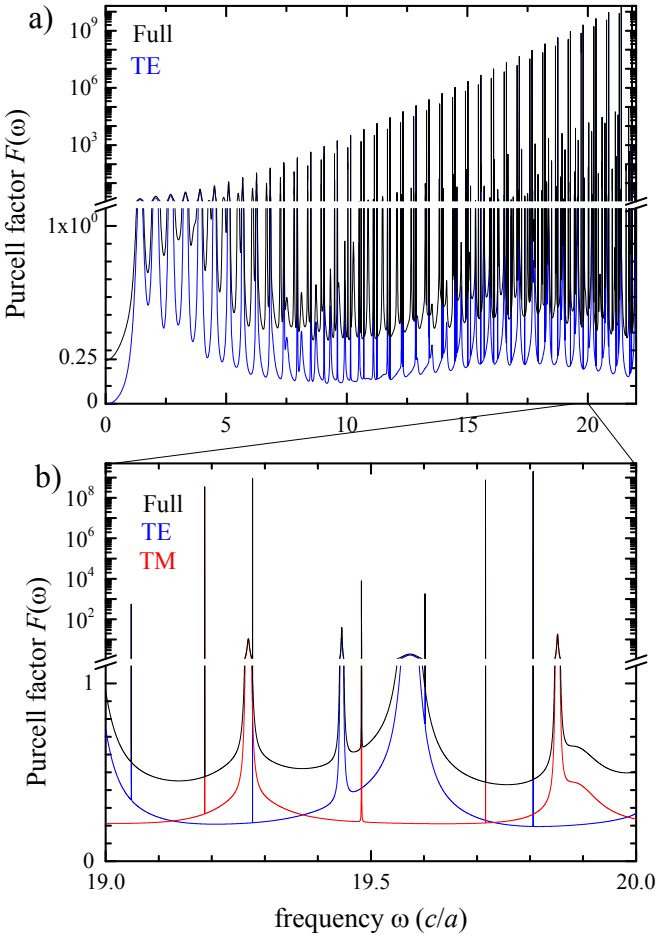

FIG. 2: (color online) Purcell factor versus transition frequency ω calculated by Eq. (9) using modes with $l < 38$ and $|\omega_n| < 40c/a$. The dipole is placed at $|\mathbf{r}_d| = 0.9a$ as in Fig. 1, and the PF is averaged over the polarization directions. a) Full PF (black line) and partial PF for TE modes only (blue line). b) zoom of a), showing additionally the partial PF for TM modes (red line).

modes with the angular momentum $l = 7$ and transverse electric (TE) polarization, summed over the degenerate states with azimuthal number $m = -l, \ldots, l$, is shown in Fig. 1a. The modes can be classified as leaky modes, whispering gallery modes (WGMs), and Fabry-Pérot (FP) modes, as indicated. The chosen dipole position is close to the field maximum of the fundamental WGM, therefore its mode volume is small and essentially real. With increasing mode order going into the FP modes, the mode volume oscillates as the field maxima and minima move across the dipole position. Interestingly, the phase of the mode volume rotates accordingly, yielding negative mode volumes at the positions of the field minima, at which the mode field is imaginary. This also elucidates that the radiative decay into the modes is not a simple superposition of Lorentzian lines describing independent channels, but shows interference. This can actually be expected, as modes of equal l, m, and polarization couple into the same outgoing loss channel.

The resulting partial PF for $l = 7$ TE modes (see Fig. 1b) is dominated by the $n = 1$ WGM providing on resonance a PF of about 20. The complex mode volume leads to non-Lorentzian features in the spectrum, due to the mode interference. The total contribution to the PF of all modes for each loss channel (for spherical symmetry all modes with equal m, l, and polarization) is strictly positive, as expected. To exemplify the issues with the normalization Eq. (4) suggested by Kristensen *et al.*, we

show the resulting PF for two finite integration volumes given by spheres of radius $R = 2a$ and $R = 30a$. The observed deviation, which is increasing with R, is showing an underestimation of the contribution of leaky and FP modes. The PF also shows negative values which are unphysical. Taking the limit $R \to \infty$, the mode volume diverges exponentially, according to Eq. (4), see Fig. S1 in the SM, – this is also true for high-Q modes but commences at larger R – so that the PF vanishes. In metallic resonators the modes have generally low Q-factors, yielding a fast exponential growth (see Fig. S2) and large errors of Eq. (4) for any $\mathcal{V}$.

To explicitly show the validity of the spectral representation and the resulting PF based on the exact normalization, we have calculated the PF using all relevant modes, i.e. taking into account both TE and transverse magnetic (TM) polarizations and summing over all significant values of l and m. Examples of mode volumes and partial PFs for the TM modes are shown in the Figs. S3 and S4 of the SM for two different directions of the dipole. The resulting PF for a dipole at a distance $0.9a$ from the center of the sphere averaged over its polarization directions is shown in Fig. 2, with partial PFs demonstrated separately for TE modes in Figs. 2(a) and 2(b) and for TM modes in Fig. 2(b). In the low-frequency limit the well known static value of the field reduction by a factor of $3/(2+\varepsilon)$ inside a dielectric sphere in vacuum is reproduced giving $F(0) = 0.25$. We have verified our result by using the analytic form of the GF of a sphere in terms of linearly independent solutions of a second-order homogeneous differential equation. The spectral zoom in Fig. 2(b) allows to see the extremely sharp WGM lines on top of much wider resonances and their separation into TE and TM modes. Purcell factors up to 10^{10} are found in resonance to WGM of similarly high Q-factors.

In conclusion, we have provided a general exact analytic form of the normalization of eigenmodes in an arbitrary open optical resonator, which rectifies a normalization expression previously believed to be valid. This quantity is of key importance for the electromagnetic theory, as it determines the spectral representation of the dyadic Green's function of Maxwell's wave equation, which can be used for calculation of any observable, such as scattering and extinction cross-sections and local density of states. We focussed in the present work on the consequences for the determination of the mode volume and the Purcell factor, which is a cornerstone for cavity quantum electrodynamics and nanoplasmonics. Further fundamental developments enabled by this exact normalization are to be expected, as exemplified by the rigorous perturbation theory in electrodynamics called the resonant-state expansion [8–12].

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Supplementary Material

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I. SPONTANEOUS EMISSION OF A QUANTUM DIPOLE IN AN ARBITRARY DIELECTRIC SYSTEM

The full Hamiltonian describing a quantum dipole coupled to photon states in an arbitrary open optical system is given [1] by

$$H = H_0 + V\,, \tag{S1}$$

where

$$H_0 = \sum_k \hbar\omega_k a_k^\dagger a_k + \hbar\omega_d d^\dagger d \tag{S2}$$

is the non-interacting part and

$$V = -i\sum_k (\varphi_k d^\dagger a_k - \varphi_k^* a_k^\dagger d) \tag{S3}$$

is the interaction between the dipole and photons in the rotating wave approximation. Here $a_k^\dagger$ is the photon creation operator in state k, $d^\dagger$ is the fermionic creation operator for the two-level system of the quantum dipole, with ω_d being the ground-to-excited state transition frequency, and

$$\varphi_k = \sqrt{\frac{\hbar\omega_k}{2\varepsilon_0}}\boldsymbol{\mu}\cdot\mathbf{f}_k(\mathbf{r}_d) \tag{S4}$$

is the coupling matrix element, in which $\boldsymbol{\mu}$ is the electric dipole moment of the point dipole placed at $\mathbf{r} = \mathbf{r}_d$, ε_0 is the vacuum permittivity and $\mathbf{f}_k(\mathbf{r})$ is a vector eigenfunction of the electric field of the continuum state k satisfying the Maxwell wave equation

$$-\nabla\times\nabla\times\mathbf{f}_k(\mathbf{r}) + \frac{\omega_k^2}{c^2}\hat{\boldsymbol{\epsilon}}(\mathbf{r})\mathbf{f}_k(\mathbf{r}) = 0 \tag{S5}$$

with a *real* eigen-frequency $\omega_k \geqslant 0$. The symmetric tensor $\hat{\boldsymbol{\epsilon}}(\mathbf{r})$ of the dielectric constant describes the open optical system under study and for simplicity is assumed here to be frequency-independent.

Following Glauber [1], we consider the Schrödinger equation describing the full system ($\hbar = 1$ is used below for brevity of notations),

$$i\frac{d}{dt}|\Phi(t)\rangle = H|\Phi(t)\rangle\,, \tag{S6}$$

and take its formal solution in the form

$$|\Phi(t)\rangle = e^{-iHt}|\Phi(0)\rangle\,, \tag{S7}$$

where $|\Phi(t)\rangle$ is the wave function of the dipole-photon system. We are interested in the probability for the dipole to stay in the excited state and calculate the probability amplitude in the following way:

$$\alpha(t) = \langle\Phi(0)|\Phi(t)\rangle = \langle 0|de^{-iHt}d^\dagger|0\rangle = \langle 0|d(t)U(t)d^\dagger|0\rangle\,. \tag{S8}$$

In the above equation, the dipole moment operator is written in the interaction representation, $d(t) = e^{iH_0t}de^{-iH_0t}$. We have also assumed that in the initial state, the photon subsystem is in its ground state and the quantum dipole is in its excited state, i.e. $|\Phi(0)\rangle = d^\dagger|0\rangle$, where $|0\rangle$ is the ground state of the full system. The evolution operator $U(t) = e^{iH_0t}e^{-iHt}$ satisfies the equation

$$i\frac{dU(t)}{dt} = V(t)U(t)\,, \tag{S9}$$

where $V(t) = e^{iH_0t}Ve^{-iH_0t}$. Its solution can be written as an infinite perturbation series

$$U(t) = 1 + (-i)\int_0^t V(t_1)dt_1 + (-i)^2\int_0^t V(t_1)dt_1\int_0^{t_1} V(t_2)dt_2 + \ldots \tag{S10}$$

To calculate $\alpha(t)$, we evaluate

$$\begin{aligned} V(t_1)V(t_2)d^\dagger|0\rangle &= -e^{iH_0t_1}\sum_{k_1}(\varphi_{k_1}d^\dagger a_{k_1} - \varphi_{k_1}^* a_{k_1}^\dagger d)\\ &\times e^{-iH_0(t_1-t_2)}\sum_{k_2}(\varphi_{k_2}d^\dagger a_{k_2} - \varphi_{k_2}^* a_{k_2}^\dagger d)e^{-iH_0t_2}d^\dagger|0\rangle\\ &= e^{i\omega_d(t_1-t_2)}\sum_k |\varphi_k|^2 e^{-i\omega_k(t_1-t_2)}d^\dagger|0\rangle\,. \end{aligned} \tag{S11}$$

Then for $t > 0$, $\alpha(t)$ can be written in the form of an integral equation:

$$\alpha(t) = e^{-i\omega_d t} - \sum_k |\varphi_k|^2\int_0^t dt_1\int_0^{t_1} dt_2 \times e^{-i\omega_d(t-t_1)}e^{-i\omega_k(t_1-t_2)}\alpha(t_2) \tag{S12}$$

which can be solved explicitly in the Fourier space:

$$\tilde{\alpha}(\omega) = \frac{-i}{\omega - \omega_d - \Sigma(\omega)}\,, \tag{S13}$$

where $\tilde{\alpha}(\omega)$ is the time Fourier transform of $\alpha(t)$, and the self-energy $\Sigma(\omega)$ is given by a formula

$$\Sigma(\omega) = \frac{1}{\hbar^2}\sum_k \frac{|\varphi_k|^2}{\omega - \omega_k + i\delta}\,, \tag{S14}$$

in which $\delta \to 0_+$ and $\hbar$ has been restored. Note that the problem described by Eqs. (S1)–(S3) is the famous exactly solvable Fano-Anderson problem. Indeed, owing to the bilinear form of the interaction Eq. (S3) the exact perturbation series for the self-energy ends in first order [2].

Let us express the self-energy $\Sigma(\omega)$ in terms of the dyadic GF of Maxwell's wave equation. The full time-dependent GF $\hat{\boldsymbol{\mathcal{G}}}(\mathbf{r},\mathbf{r}';t-t')$ satisfies the equation

$$-\nabla\times\nabla\times\hat{\boldsymbol{\mathcal{G}}}-\frac{\hat{\boldsymbol{\epsilon}}(\mathbf{r})}{c^2}\frac{\partial^2\hat{\boldsymbol{\mathcal{G}}}}{\partial t^2}=\hat{\mathbf{1}}\delta(\mathbf{r}-\mathbf{r}')\delta(t-t') \quad \text{(S15)}$$

and has the following explicit form in terms of the continuum eigenstates, the solutions of Eq. (S5):

$$\hat{\boldsymbol{\mathcal{G}}}(\mathbf{r},\mathbf{r}';t-t')=\frac{c^2}{i}\sum_k\frac{\mathbf{f}_k(\mathbf{r})\otimes\mathbf{f}_k^*(\mathbf{r}')}{2\omega_k}\,e^{-i\omega_k|t-t'|}\,. \quad \text{(S16)}$$

Note that substituting Eq. (S16) into Eq. (S15) and using Eq. (S5) results in the closure relation for the continuum eigenstates,

$$\hat{\boldsymbol{\epsilon}}(\mathbf{r})\sum_k\mathbf{f}_k(\mathbf{r})\otimes\mathbf{f}_k^*(\mathbf{r}')=\hat{\mathbf{1}}\delta(\mathbf{r}-\mathbf{r}')\,. \quad \text{(S17)}$$

Fourier transforming the GF given by Eq. (S16) versus $t-t'$ we obtain

$$\hat{\mathbf{G}}(\mathbf{r},\mathbf{r}';\omega)=c^2\sum_k\frac{\mathbf{f}_k(\mathbf{r})\otimes\mathbf{f}_k^*(\mathbf{r}')}{\omega^2-\omega_k^2+i\delta}\,. \quad \text{(S18)}$$

Then, using Eq. (S4) for a positive frequency ω we find

$$\begin{aligned} I(\mathbf{r}_d,\omega)&\equiv\boldsymbol{\mu}\cdot\mathrm{Im}\,\hat{\mathbf{G}}(\mathbf{r}_d,\mathbf{r}_d;\omega)\boldsymbol{\mu}\\ &=-\frac{\pi\varepsilon_0c^2}{\hbar}\sum_k\frac{|\varphi_k|^2}{\omega_k^2}\delta(\omega-\omega_k)\,, \end{aligned} \quad \text{(S19)}$$

that allows us to express the self-energy in terms of the projection $I(\mathbf{r},\omega)$ of the GF tensor:

$$\begin{aligned} \Sigma(\omega)&=\frac{1}{\hbar^2}\sum_k\int_0^\infty\frac{|\varphi_k|^2\delta(\omega'-\omega_k)d\omega'}{\omega-\omega'+i\delta}\\ &=-\frac{1}{\pi\varepsilon_0c^2\hbar}\int_0^\infty\frac{I(\mathbf{r}_d,\omega')\omega'^2}{\omega-\omega'+i\delta}d\omega'\,. \end{aligned} \quad \text{(S20)}$$

For the GF of a homogeneous medium with the dielectric constant ε we have

$$\mathrm{Im}\,\hat{\mathbf{G}}^0(\mathbf{r},\mathbf{r};\omega)=-\frac{\sqrt{\varepsilon}\omega}{6\pi c}\hat{\mathbf{1}} \quad \text{(S21)}$$

as shown below, such that the integral in Eq. (S20) diverges for large ω', which is the well known divergence problem of the Lamb shift, usually treated by introducing a frequency cut-off. For an inhomogeneous open optical system this integral is, however, convergent. Indeed, using the spectral representation of the GF in terms of resonant states (RSs) with *complex* eigenfrequencies ω_n [3, 4]

$$\hat{\mathbf{G}}(\mathbf{r},\mathbf{r}';\omega)=c^2\sum_n\frac{\mathbf{E}_n(\mathbf{r})\otimes\mathbf{E}_n(\mathbf{r}')}{2\omega(\omega-\omega_n)} \quad \text{(S22)}$$

(see also Sec. II), we obtain for any $\mathbf{r}$ inside the system

$$I(\mathbf{r},\omega)=\frac{c^2}{2\omega}\mathrm{Im}\sum_n\frac{g_n^2(\mathbf{r})}{\omega-\omega_n}\,, \quad \text{(S23)}$$

where

$$g_n(\mathbf{r})=\boldsymbol{\mu}\cdot\mathbf{E}_n(\mathbf{r})\,. \quad \text{(S24)}$$

We note that RSs contribute to Eq. (S22) in pairs: Each RS n with the eigenfrequency ω_n and electric field eigenfunction $\mathbf{E}_n(\mathbf{r})$ has a counterpart $-n$ with $\omega_{-n}=-\omega_n^*$ and $\mathbf{E}_{-n}(\mathbf{r})=\mathbf{E}_n^*(\mathbf{r})$. Their joint contribution to Eq. (S23) is given by

$$\begin{aligned} &\mathrm{Im}\left[\frac{g_n^2(\mathbf{r})}{\omega-\omega_n}+\frac{g_n^{*2}(\mathbf{r})}{\omega+\omega_n^*}\right]\\ &=\frac{A_n''(\omega-\omega_n')+A_n'\omega_n''}{(\omega-\omega_n')^2+\omega_n''^2}+\frac{-A_n''(\omega+\omega_n')+A_n'\omega_n''}{(\omega+\omega_n')^2+\omega_n''^2}\,, \end{aligned} \quad \text{(S25)}$$

where $g_n^2(\mathbf{r})=A_n'+iA_n''$ and $\omega_n=\omega_n'+i\omega_n''$. Therefore $I(\mathbf{r},\omega)\propto 1/\omega^3$ at $\omega\to\infty$ and the integral in Eq. (S20) converges.

For small values of ω, it is more practical to use a different form of the GF [3, 4]

$$\hat{\mathbf{G}}(\mathbf{r},\mathbf{r}';\omega)=c^2\sum_n\frac{\mathbf{E}_n(\mathbf{r})\otimes\mathbf{E}_n(\mathbf{r}')}{2\omega_n(\omega-\omega_n)}\,, \quad \text{(S26)}$$

which follows from Eq. (S22) and the sum rule (see Sec. II)

$$\sum_n\frac{\mathbf{E}_n(\mathbf{r})\otimes\mathbf{E}_n(\mathbf{r}')}{\omega_n}=0\,. \quad \text{(S27)}$$

Then $I(\mathbf{r},\omega)$ can then be written as:

$$I(\mathbf{r},\omega)=\frac{c^2}{2}\mathrm{Im}\sum_n\frac{g_n^2(\mathbf{r})}{\omega_n(\omega-\omega_n)}\,, \quad \text{(S28)}$$

and the contribution of the pair of poles takes the form

$$\begin{aligned} &\mathrm{Im}\left[\frac{g_n^2(\mathbf{r})}{\omega_n(\omega-\omega_n)}-\frac{g_n^{*2}(\mathbf{r})}{\omega_n^*(\omega+\omega_n^*)}\right]\\ &=\frac{B_n''(\omega-\omega_n')+B_n'\omega_n''}{(\omega-\omega_n')^2+\omega_n''^2}-\frac{-B_n''(\omega+\omega_n')+B_n'\omega_n''}{(\omega+\omega_n')^2+\omega_n''^2}\,, \end{aligned} \quad \text{(S29)}$$

where $g_n^2(\mathbf{r})/\omega_n=B_n'+iB_n''$, so that $I(\mathbf{r},\omega)\propto\omega$ in the limit $\omega\to 0$.

Note that for the poles of the GF on the imaginary ω-axis which do not have counterparts, $g_n^2(\mathbf{r})$ is real, and both low- and high-frequency asymptotics of $I(\mathbf{r},\omega)$ obtained above are preserved. Moreover, for the same reason, static modes (having $\omega_n=0$) do not contribute to the spontaneous emission, as the corresponding term of the GF is purely real (the modes are localized).

Using Eqs. (S23) or (S28), $\Sigma(\omega)$ can be calculated analytically for any finite number of RSs, thus providing

direct access to the analytic continuation of $\tilde{\alpha}(\omega)$ into the complex ω plane and to its pole structure. Owing to the causality principle, $\tilde{\alpha}(\omega)$ has poles only in the lower half plane, which results in the following expression for the probability amplitude in the time domain:

$$\alpha(t) = \theta(t) \sum_j c_j e^{-i(\omega_d + \delta\omega_j)t - \gamma_j t}\,, \tag{S30}$$

where

$$\frac{1}{c_j} = 1 - \left.\frac{d\Sigma(\omega)}{d\omega}\right|_{\omega=\omega_j} \tag{S31}$$

and $\omega_j = \omega_d + \delta\omega_j - i\gamma_j$ are the poles of $\tilde{\alpha}(\omega)$. Such an analysis is important for the strong coupling regime. In the weak coupling regime instead, $\Sigma(\omega)$ can be considered as a small correction, and $\tilde{\alpha}(\omega)$ can be treated in the single-pole approximation leading to

$$\alpha(t) = \theta(t) e^{-i(\omega_d + \delta\omega)t - \gamma t}\,, \tag{S32}$$

where $\delta\omega - i\gamma = \Sigma(\omega_d)$ is the self-energy correction to the pole of GF of the dipole, calculated "on-shell", i.e. at $\omega = \omega_d$. The Lamb shift $\delta\omega$ and the spontaneous emission rate γ then take the following explicit form

$$\delta\omega = \frac{1}{\pi\varepsilon_0 c^2 \hbar} \int_0^\infty \frac{I(\mathbf{r}_d, \omega)\omega^2 d\omega}{\omega - \omega_d}\,, \tag{S33}$$

$$\gamma(\omega_d) = -\frac{\omega_d^2}{\varepsilon_0 c^2 \hbar} I(\mathbf{r}_d, \omega_d), \tag{S34}$$

where the principal value integral is introduced in Eq. (S33). Equations (S32)–(S34) are know in the literature as the Weisskopf-Wigner approximation [5].

Let us check that Eq. (S34) produces the correct expression for the spontaneous emission rate in the case of a homogeneous dielectric medium. The GF of the free space satisfies the equation

$$-\nabla\times\nabla\times\hat{\mathbf{G}}^0(\mathbf{r},\mathbf{r}';\omega) + k^2\hat{\mathbf{G}}^0(\mathbf{r},\mathbf{r}';\omega) = \hat{\mathbf{1}}\delta(\mathbf{r}-\mathbf{r}')\,, \tag{S35}$$

where $k^2 = \varepsilon\omega^2/c^2$ and ε is the dielectric constant of the medium. The solution of Eq. (S35) has the form [6]:

$$\hat{\mathbf{G}}^0(\mathbf{r},\mathbf{r}';\omega) = -\left(\hat{\mathbf{1}} + \frac{1}{k^2}\nabla\otimes\nabla\right)\frac{e^{ik|\mathbf{r}-\mathbf{r}'|}}{4\pi|\mathbf{r}-\mathbf{r}'|}\,, \tag{S36}$$

or, more explicitly [7],

$$\hat{\mathbf{G}}^0(\mathbf{r},\mathbf{r}';\omega) = C\,\hat{\mathbf{1}} + D\,\frac{\mathbf{b}\otimes\mathbf{b}}{b^2}\,, \tag{S37}$$

where $\mathbf{b} = \mathbf{r} - \mathbf{r}'$, $b = |\mathbf{b}|$ and

$$C = -\left(1 + \frac{ikb-1}{k^2b^2}\right)\frac{e^{ikb}}{4\pi b} = \frac{2-k^2b^2}{8\pi k^2 b^3} - i\frac{k}{6\pi} + k\mathcal{O}(kb)\,, \tag{S38}$$

$$D = -\frac{3-3ikb-k^2b^2}{k^2b^2}\frac{e^{ikb}}{4\pi b} = -\frac{6+k^2b^2}{8\pi k^2 b^3} + k\mathcal{O}(kb)\,, \tag{S39}$$

expanded up to zeroth order in kb. Taking the limit $\mathbf{r}' \to \mathbf{r}$, so that $b \to 0$, we obtain Eq. (S21) and finally

$$\gamma_0(\omega) = -\frac{\omega^2}{\varepsilon_0\hbar c^2}\boldsymbol{\mu}\cdot\mathrm{Im}\,\hat{\mathbf{G}}^0(\mathbf{r}_d,\mathbf{r}_d;\omega)\boldsymbol{\mu} = \frac{\sqrt{\varepsilon}\omega^3\mu^2}{6\pi\epsilon_0\hbar c^3}\,, \tag{S40}$$

in agreement with Ref. [8]. Note that using the spectral representation of the GF in the form of Eq. (S26), the spontaneous decay rate $\gamma(\omega)$ of an inhomogeneous open optical system also scales like ω^3 at $\omega \to 0$ [$I(\mathbf{r}_d,\omega) \propto \omega$ as demonstrated above]. This makes the Purcell factor (PF) $F(\omega) = \gamma(\omega)/\gamma_0(\omega)$ finite at $\omega \to 0$.

II. SPECTRAL REPRESENTATION OF THE GREEN'S FUNCTION AND NORMALIZATION OF RESONANT STATES

The Green's function $\hat{\mathbf{G}}(\mathbf{r},\mathbf{r}';\omega)$ of an open optical system is a tensor which satisfies Maxwell's wave equation with a delta-function source term (below $c = 1$ is used for brevity of notations),

$$-\nabla\times\nabla\times\hat{\mathbf{G}}(\mathbf{r},\mathbf{r}';\omega) + \omega^2\hat{\boldsymbol{\epsilon}}(\mathbf{r};\omega)\hat{\mathbf{G}}(\mathbf{r},\mathbf{r}';\omega) = \hat{\mathbf{1}}\delta(\mathbf{r}-\mathbf{r}')\,, \tag{S41}$$

and outgoing wave boundary conditions. Treating $\hat{\mathbf{G}}(\mathbf{r},\mathbf{r}';\omega)$ as a function of a complex ω we use the fact that the GF has a countable number of simple poles in the lower half plane at $\omega = \tilde{\omega}_n$. We further note that for $\omega \to \infty$, the GF vanishes inside the area of inhomogeneity of $\hat{\boldsymbol{\epsilon}}(\mathbf{r};\omega)$. Note that the frequency dependence of the permittivity tensor is included. Then according to the Mittag-Leffler theorem [9, 10] we can write

$$\hat{\mathbf{G}}(\mathbf{r},\mathbf{r}';\omega) = \sum_n \frac{\hat{\mathbf{R}}_n(\mathbf{r},\mathbf{r}')}{\omega - \tilde{\omega}_n}\,, \tag{S42}$$

where $\hat{\mathbf{R}}_n(\mathbf{r},\mathbf{r}')$ is the residue of the GF at $\omega = \tilde{\omega}_n$.

Now, for each RS having the eigenfrequency ω_n and the electric field $\mathbf{E}_n(\mathbf{r})$ satisfying the homogeneous Maxwell wave equation

$$-\nabla\times\nabla\times\mathbf{E}_n(\mathbf{r}) + \omega_n^2\hat{\boldsymbol{\epsilon}}(\mathbf{r};\omega_n)\mathbf{E}_n(\mathbf{r}) = 0 \tag{S43}$$

and outgoing wave boundary conditions, we introduce an analytic continuation $\mathbf{F}_n(\mathbf{r},\omega)$, such that

$$\lim_{\omega\to\omega_n}\mathbf{F}_n(\mathbf{r},\omega) = \mathbf{E}_n(\mathbf{r})\,. \tag{S44}$$

$\mathbf{F}_n(\mathbf{r},\omega)$ is defined as a solution of the inhomogeneous Maxwell wave equations

$$-\nabla\times\nabla\times\mathbf{F}_n(\mathbf{r};\omega) + \omega^2\hat{\boldsymbol{\epsilon}}(\mathbf{r};\omega)\mathbf{F}_n(\mathbf{r};\omega) = (\omega^2-\omega_n^2)\boldsymbol{\sigma}_n(\mathbf{r})\,, \tag{S45}$$

in which $\boldsymbol{\sigma}_n(\mathbf{r})$ is an arbitrary function vanishing outside the system and normalized in such a way that

$$\int_{\mathcal{V}}\mathbf{E}_n(\mathbf{r})\cdot\boldsymbol{\sigma}_n(\mathbf{r})\,d\mathbf{r} = 1\,, \tag{S46}$$

where $\mathcal{V}$ is an arbitrary simply connected volume including all the inhomogeneities of $\hat{\boldsymbol{\epsilon}}(\mathbf{r};\omega)$. In the case of degenerate modes, $\omega_m = \omega_n$ for $m \neq n$, the source $\boldsymbol{\sigma}_n(\mathbf{r})$ has to be chosen in such a way that, additionally, $\int_{\mathcal{V}} \mathbf{E}_m(\mathbf{r}) \cdot \boldsymbol{\sigma}_n(\mathbf{r})\, d\mathbf{r} = 0$. Solving Eq. (S45) with the help of the GF Eq. (S42) we obtain

$$\mathbf{F}_n(\mathbf{r};\omega) = \sum_{n'} \frac{\omega^2 - \omega_n^2}{\omega - \tilde{\omega}_{n'}} \int_{\mathcal{V}} \hat{\mathbf{R}}_{n'}(\mathbf{r},\mathbf{r}') \boldsymbol{\sigma}_n(\mathbf{r}')\, d\mathbf{r}' \,. \quad \text{(S47)}$$

Taking the limit of Eq. (S44) and using the fact that $\hat{\mathbf{G}}(\mathbf{r},\mathbf{r}';\omega)$ is a symmetric tensor, which follows from the reciprocity theorem [11], we find

$$\hat{\mathbf{R}}_n(\mathbf{r},\mathbf{r}') = \frac{\mathbf{E}_n(\mathbf{r}) \otimes \mathbf{E}_n(\mathbf{r}')}{2\omega_n} \quad \text{(S48)}$$

and $\omega_n = \tilde{\omega}_n$, leading to the spectral representation Eq. (S26). Substituting it into Eq. (S41) and using Eq. (S43) results in the closure relation

$$\sum_n \frac{\omega^2 \hat{\boldsymbol{\epsilon}}(\mathbf{r};\omega) - \omega_n^2 \hat{\boldsymbol{\epsilon}}(\mathbf{r};\omega_n)}{2\omega_n(\omega - \omega_n)} \mathbf{E}_n(\mathbf{r}) \otimes \mathbf{E}_n(\mathbf{r}') = \hat{\mathbf{1}} \delta(\mathbf{r} - \mathbf{r}') \,, \quad \text{(S49)}$$

which in the absence of frequency dispersion of $\hat{\boldsymbol{\epsilon}}(\mathbf{r})$ splits into the sum rule Eq. (S27) and a simpler closure relation

$$\frac{1}{2}\, \hat{\boldsymbol{\epsilon}}(\mathbf{r}) \sum_n \mathbf{E}_n(\mathbf{r}) \otimes \mathbf{E}_n(\mathbf{r}') = \hat{\mathbf{1}} \delta(\mathbf{r} - \mathbf{r}') \,. \quad \text{(S50)}$$

As already noted in Sec. I, combining Eq. (S26) and the sum rule Eq. (S27) leads to an alternative form of the spectral representation Eq. (S22) which was used in the resonant-state expansion (RSE) [3, 4].

The form of the GF Eq. (S22) determines the normalization of RSs which technically follows from Eq. (S46) by substituting $\boldsymbol{\sigma}_n(\mathbf{r})$ from Eq. (S45) and taking the limit $\omega \to \omega_n$ (below the argument $\mathbf{r}$ is omitted for brevity):

$$\begin{aligned}
1 &= \int_{\mathcal{V}} d\mathbf{r}\, \mathbf{E}_n \cdot \boldsymbol{\sigma}_n \\
&= \lim_{\omega\to\omega_n} \int_{\mathcal{V}} d\mathbf{r}\, \mathbf{E}_n \cdot \frac{-\nabla\times\nabla\times\mathbf{F}_n + \omega^2 \hat{\boldsymbol{\epsilon}}(\omega)\mathbf{F}_n}{\omega^2 - \omega_n^2} \\
&\quad - \lim_{\omega\to\omega_n} \int_{\mathcal{V}} d\mathbf{r}\, \mathbf{F}_n \cdot \frac{-\nabla\times\nabla\times\mathbf{E}_n + \omega_n^2 \hat{\boldsymbol{\epsilon}}(\omega_n)\mathbf{E}_n}{\omega^2 - \omega_n^2} \\
&= \lim_{\omega\to\omega_n} \int_{\mathcal{V}} d\mathbf{r}\, \mathbf{F}_n \cdot \frac{\omega^2 \hat{\boldsymbol{\epsilon}}(\omega) - \omega_n^2 \hat{\boldsymbol{\epsilon}}(\omega_n)}{\omega^2 - \omega_n^2} \mathbf{E}_n \\
&\quad + \lim_{\omega\to\omega_n} \frac{\int_{\mathcal{V}} (\mathbf{F}_n \cdot \nabla\times\nabla\times\mathbf{E}_n - \mathbf{E}_n \cdot \nabla\times\nabla\times\mathbf{F}_n) d\mathbf{r}}{\omega^2 - \omega_n^2} \\
&= \int_{\mathcal{V}} d\mathbf{r}\, \mathbf{E}_n \cdot \left.\frac{\partial(\omega^2 \hat{\boldsymbol{\epsilon}}(\omega))}{\partial(\omega^2)}\right|_{\omega=\omega_n} \mathbf{E}_n \\
&\quad + \lim_{\omega\to\omega_n} \frac{\oint_{S_{\mathcal{V}}} dS \left(\mathbf{E}_n \cdot \dfrac{\partial \mathbf{F}_n}{\partial s} - \mathbf{F}_n \cdot \dfrac{\partial \mathbf{E}_n}{\partial s} \right)}{\omega^2 - \omega_n^2} \,, \quad \text{(S51)}
\end{aligned}$$

where after using some vector algebra we have applied the divergence theorem to convert a volume integral into a surface integral over the closed surface $S_{\mathcal{V}}$, the boundary of $\mathcal{V}$, with $\partial/\partial s$ denoting the directional derivative normal to this surface.

For any surface $S_{\mathcal{V}}$, the limit in the last term in Eq. (S51) can be evaluated explicitly by using the functional dependence of the electric field outside the system, where $\hat{\boldsymbol{\epsilon}}(\mathbf{r}) = \hat{\mathbf{1}}$ up to a scalar constant. For any mode with $\omega_n \neq 0$, the wave function of the RS is given by $\mathbf{E}_n(\mathbf{r}) = \mathbf{Q}_n(\omega_n \mathbf{r})$, where $\mathbf{Q}_n(\mathbf{q})$ is a vector function satisfying the equation

$$\nabla_{\mathbf{q}} \times \nabla_{\mathbf{q}} \times \mathbf{Q}_n(\mathbf{q}) = \mathbf{Q}_n(\mathbf{q}) \quad \text{(S52)}$$

and the proper boundary conditions at system interfaces and at $\mathbf{q} \to \infty$. The analytic continuation of $\mathbf{E}_n(\mathbf{r})$ can therefore be taken in the form

$$\mathbf{F}_n(\mathbf{r},\omega) = \mathbf{Q}_n(\omega\mathbf{r}) \,. \quad \text{(53)}$$

Using the Taylor expansion about the point $\omega = \omega_n$,

$$\begin{aligned}
\mathbf{F}_n(\mathbf{r},\omega) &\approx \mathbf{Q}_n(\omega_n\mathbf{r}) + (\omega - \omega_n) r \left.\frac{\partial \mathbf{Q}_n(\omega\mathbf{r})}{\partial(\omega r)}\right|_{\omega=\omega_n} \\
&= \mathbf{E}_n(\mathbf{r}) + \frac{\omega - \omega_n}{\omega_n} r \frac{\partial \mathbf{E}_n(\mathbf{r})}{\partial r} \,, \quad \text{(S54)}
\end{aligned}$$

where $r = |\mathbf{r}|$ is the radius in the spherical coordinates, and substituting Eq. (S54) into Eq. (S51) we obtain

$$\begin{aligned}
1 = &\int_{\mathcal{V}} d\mathbf{r}\, \mathbf{E}_n \cdot \left.\frac{\partial(\omega^2 \hat{\boldsymbol{\epsilon}}(\omega))}{\partial(\omega^2)}\right|_{\omega=\omega_n} \mathbf{E}_n \quad \text{(S55)} \\
&+ \frac{c^2}{2\omega_n^2} \oint_{S_{\mathcal{V}}} dS \left[\mathbf{E}_n \cdot \frac{\partial}{\partial s} r \frac{\partial \mathbf{E}_n}{\partial r} - r \frac{\partial \mathbf{E}_n}{\partial r} \cdot \frac{\partial \mathbf{E}_n}{\partial s} \right]
\end{aligned}$$

where the speed of light is restored. In the absence of dispersion the first integral in the normalization Eq. (S55) is simplified to $\int_{\mathcal{V}} d\mathbf{r}\, \mathbf{E}_n \cdot \hat{\boldsymbol{\epsilon}} \mathbf{E}_n$, yielding Eq. (8) of the main text.

We note that the normalization of static modes ($\omega_n = 0$) is different and has been treated in Ref. [4]. They do not contribute to the radiative decay and thus are not further considered here.

III. NORMALIZATION BY KRISTENSEN ET AL.

Following Leung *et al.* [12], Kristensen *et al.* [13] have introduced a normalization of RSs in the form of Eq. (4) of the main text. We found that this normalization is only correct for so-called s-waves, i.e. $l = 0$ modes of a spherically symmetric system, where l is the orbital quantum number. However, owing to the vectorial nature of the electromagnetic field, $l = 0$ eigenmodes do not exist, so that Eq. (4) is incorrect for all modes in electrodynamics.

We illustrate this finding for transverse-electric (TE) modes of a dielectric sphere. We compare the approximate mode volume V_n^{ap}, calculated using Eq. (4), with

the correct one, V_n, calculated using the normalization Eq. (8), by considering the relation

$$V_n^{\rm ap} = V_n N_n(R)\,, \tag{S56}$$

in which $N_n(R)$ is the factor between the two volumes. It is given by the sum of the volume and surface normalization integrals in Eq. (4):

$$N_n(R) = I_n(R) + S_n(R) \tag{S57}$$

with

$$I_n(R) = \int_{\mathcal{V}_R} \varepsilon(\mathbf{r})\mathbf{E}_n^2(\mathbf{r})d\mathbf{r} \tag{S58}$$

and

$$S_n(R) = \frac{i}{2k_n}\oint_{S_R} \mathbf{E}_n^2(\mathbf{r})dS\,. \tag{S59}$$

Here $\mathcal{V}_R$ is the volume of a sphere of radius R, S_R is its surface and $k_n = \omega_n/c$ is the RS wave number. The normalization Eq. (4) requires that the factor $N_n(R)$ is unity in the limit $R\to\infty$. To illustrate the error of Eq. (4), we calculate $N_n(R)$ for the correctly normalized $\mathbf{E}_n(\mathbf{r})$ for finite R and in the limit $R\to\infty$.

For a dielectric sphere of radius a in vacuum, described by the dielectric constant

$$\varepsilon(r) = \begin{cases} n_r^2 & \text{for } r\leqslant a \\ 1 & \text{for } r>a\,, \end{cases} \tag{S60}$$

the eigenfunctions of the TE modes normalized via Eq. (8) have the form (in spherical polar coordinates) [4]:

$$\mathbf{E}_n^{\rm TE}(\mathbf{r}) = A_l^{\rm TE} R_l(r,k_n)\begin{pmatrix} 0 \\ \dfrac{1}{\sin\theta}\dfrac{\partial}{\partial\varphi}Y_{lm}(\Omega) \\ -\dfrac{\partial}{\partial\theta}Y_{lm}(\Omega) \end{pmatrix}, \tag{S61}$$

where $Y_{lm}(\Omega)$ are the spherical harmonics,

$$R_l(r,k) = \begin{cases} j_l(n_r kr)/j_l(n_r ka) & \text{for } r\leqslant a \\ h_l(kr)/h_l(ka) & \text{for } r>a\,, \end{cases} \tag{S62}$$

$j_l(z)$ and $h_l(z)\equiv h_l^{(1)}(z)$ are, respectively, the spherical Bessel and Hankel functions of first kind,

$$A_l^{\rm TE} = \sqrt{\frac{2}{l(l+1)a^3(n_r^2-1)}} \tag{S63}$$

are normalization constants and k_n are the solutions of the secular equation

$$\frac{n_r j_{l+1}(n_r k_n a)}{j_l(n_r k_n a)} = \frac{h_{l+1}(k_n a)}{h_l(k_n a)}\,. \tag{S64}$$

Using these properties, we evaluate the volume and surface normalization integrals for $R\geqslant a$ as

$$\begin{aligned} I_n(R) &= l(l+1)A_l^2\int_0^R R_l^2(r,k_n)\varepsilon(r)r^2dr \\ &= \frac{2}{a^3(n_r^2-1)}\left[n_r^2\int_0^a \frac{j_l^2(n_r k_n r)}{j_l^2(n_r k_n a)}r^2dr + \int_a^R \frac{h_l^2(k_n r)}{h_l^2(k_n a)}r^2dr\right] \\ &= 1 + \frac{(R/a)^3}{n_r^2-1}\frac{h_l^2(k_n R)}{h_l^2(k_n a)}\left[1 - \frac{h_{l-1}(k_n R)h_{l+1}(k_n R)}{h_l^2(k_n R)}\right] \end{aligned} \tag{S65}$$

and

$$S_n(R) = \frac{iR^2}{2k_n}l(l+1)A_l^2R_l^2(R,k_n) = \frac{i}{k_n R}\frac{(R/a)^3}{n_r^2-1}\frac{h_l^2(k_n R)}{h_l^2(k_n a)}\,. \tag{S66}$$

and consequently find

$$N_n(R) = 1 + \frac{1}{n_r^2-1}\left(\frac{R}{a}\right)^3\frac{h_l^2(k_n R)}{h_l^2(k_n a)}Q_n(R)\,, \tag{S67}$$

where

$$Q_n(R) = 1 - \frac{h_{l-1}(k_n R)h_{l+1}(k_n R)}{h_l^2(k_n R)} + \frac{i}{k_n R}\,. \tag{S68}$$

To investigate the behaviour of $N_n(R)$ for large $k_n R$, we use the asymptotic formula for $h_l(z)$ at large arguments. We find that the 0th- and 1st-order terms in $1/(k_n R)$ are vanishing in $Q_n(R)$, so that

$$Q_n(R) = \frac{C_l}{(k_n R)^2} + \mathcal{O}\left(\frac{1}{(k_n R)^3}\right), \tag{S69}$$

with non-vanishing constants C_l. Consequently we find

$$\frac{V_n^{\rm ap}}{V_n} = N_n(R) = 1 + \tilde{C}_l\frac{e^{2ik_n(R-a)}}{Rak_n^2} \tag{S70}$$

with $\tilde{C}_l = C_l/(n_r^2-1) + \mathcal{O}(1/(k_n R))$. Similarly, for the normalization without surface term, Eq. (3) of the main text, we find

$$\frac{V_n^{\rm ap'}}{V_n} = I_n(R) = 1 + \tilde{D}_l\frac{Re^{2ik_n(R-a)}}{a}\,, \tag{S71}$$

where $\tilde{D}_l = D_l + \mathcal{O}(1/(k_n R))$ with non-vanishing R-independents constants D_l. Clearly, Eq. (4) brings an improvement compared to Eq. (3) – the second term in Eq. (S70) is decreasing with R for some high-Q modes, such as whispering gallery modes (WGMs), for which $|e^{2ik_n(R-a)}|\approx 1$ up to rather large R. However both normalizations diverge for $R\to\infty$ due to the exponential factor $e^{2ik_n(R-a)}$.

This is exemplified in Figs. S1(b) and S2(b) where the relative errors in the mode volume, $V_n^{\rm ap}/V_n - 1$, are shown for several RSs of a dielectric and metal sphere, respectively, with corresponding eigenfrequencies given

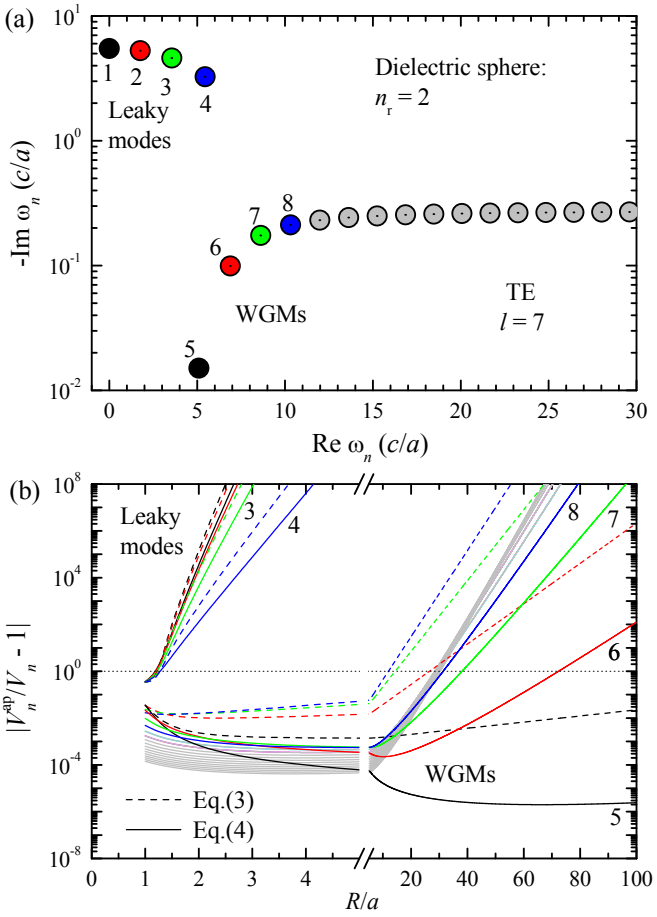

FIG. S1: (a) Frequencies of $l = 7$ TE modes (RSs) of a dielectric sphere in vacuum, with permittivity $\epsilon = 4$ and radius a. (b) Relative error of the approximate mode volume $|V_n^{\rm ap}/V_n - 1|$ as function of the radius R of the sphere of integration, for the modes shown in (a). $V_n^{\rm ap}$ is calculated using Eq. (3), having no surface term, and Eq. (4), having the incorrect surface term used in the literature [12, 13].

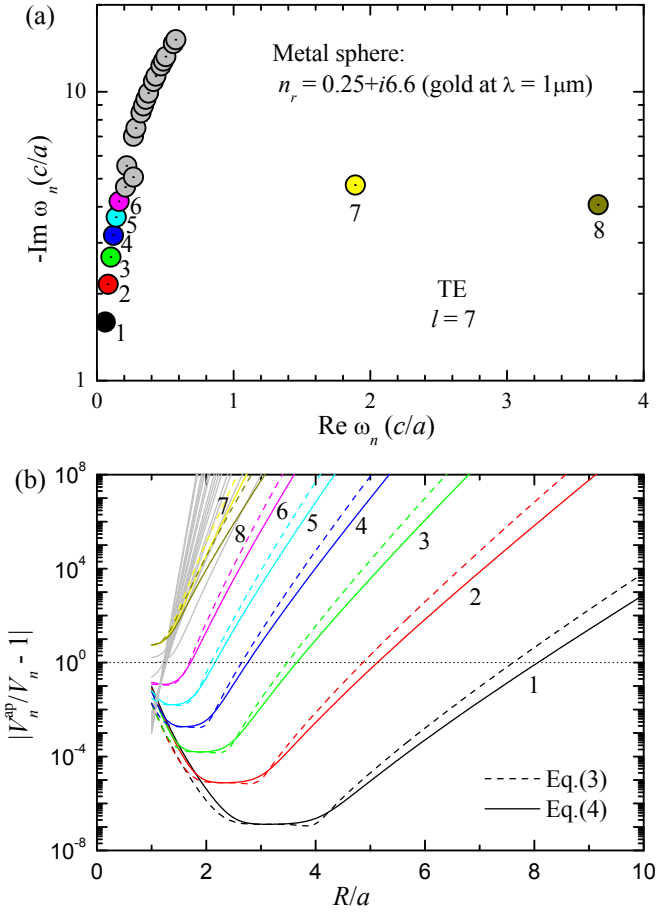

FIG. S2: As Fig. S1, but for a sphere with a fixed complex dielectric constant $\epsilon = -43.5 + i3.33$, equal to that of gold at a light wavelength of $\lambda = 1\,\mu$m.

in Figs. S1(a) and S2(a). In Fig. S1(b), the strongest deviation and exponentially growing errors are seen for leaky modes already for small values of R. For WGMs the errors can be small up to rather large R, showing an apparent convergence, in agreement with the analytic treatment given above, and the advantage of using Eq. (4) versus Eq. (3) is clearly observed. Nevertheless, the error diverges also for WGMs in the limit $R \to \infty$, in agreement with the asymptotics given by Eqs. (S70) and (S71). Moving to the metal sphere we observe that the modes have typically a low Q, such that the exponential divergence of the error with R is more pronounced. For some low-frequency modes (labeled 1-5) the error initially decays exponentially up to a finite R where the error is minimized. We note that this is observed both for Eq. (4) and Eq. (3), indicating that the surface term is not the relevant aspect here, and we find that it actually increases the error at small R. These states are quasi-bound states in the metal sphere which are evanescent close to the sphere due to the angular momentum, similar to WGMs. At $R \gtrsim lc/|\omega_n|$, they become propagating, and the error recovers the expected exponential divergence.

The correct normalization Eq. (8) can be analyzed in a similar way. It consists of two terms, $I_n(R)$ and $J_n(R)$, where

$$\begin{aligned} J_n(R) =& \frac{1}{2k_n^2}\oint_{S_R} dS\left[\mathbf{E}_n\cdot\frac{\partial}{\partial r}r\frac{\partial\mathbf{E}_n}{\partial r} - r\left(\frac{\partial\mathbf{E}_n}{\partial r}\right)^2\right] \\ =& \frac{1}{2k_n^2}\int d\Omega\left[\frac{1}{\sin^2\theta}\left(\frac{\partial Y_{lm}}{\partial\varphi}\right)^2 + \left(\frac{\partial Y_{lm}}{\partial\varphi}\right)^2\right] \\ & \times A_l^2R^2\left[R_l\frac{\partial}{\partial r}r\frac{\partial R_l}{\partial r} - r\left(\frac{\partial R_l}{\partial r}\right)^2\right]_{r=R} \\ =& \frac{R^3}{a^3}\frac{zh_l(z)h_l'(z) + z^2h_l(z)h_l''(z) - z^2h_l'^2(z)}{(n_r^2-1)h_l^2(k_na)z^2} \end{aligned} \tag{S72}$$

with $z = k_n R$. We thus obtain

$$I_n(R) + J_n(R) = 1 + \frac{R^3}{a^3(n_r^2-1)} \frac{p_l(z)}{h_l^2(k_n a) z^2}\,, \quad \text{(S73)}$$

where

$$p_l(z) = z h_l(z) h_l'(z) + z^2 h_l(z) h_l''(z) - z^2 h_l'^2(z) + z^2 h_l^2(z) - z^2 h_{l-1}(z) h_{l+1}(z) = 0\,, \quad \text{(S74)}$$

according to Bessel's equation and recursive relations for Hankel functions [14] following from it. This confirms that Eq. (8) provides the exact normalization condition $I_n(R) + J_n(R) = 1$, independent of R.

IV. COMPARISON WITH SAUVAN ET AL.

In the normalization suggested by Sauvan *et al.* [15] the electric field $\mathbf{E}$ of a RS (we drop here the index n) is normalized in such a way that

$$I_1 + I_2 = 1\,, \quad \text{(S75)}$$

where

$$I_1 = \int_{\mathcal{V}_1} d\mathbf{r}\, \mathbf{E} \cdot \frac{\partial(\omega \hat{\boldsymbol{\epsilon}}(\omega))}{\partial \omega} \mathbf{E} - \int_{\mathcal{V}_1} d\mathbf{r}\, \mathbf{H} \cdot \frac{\partial(\omega \hat{\boldsymbol{\mu}}(\omega))}{\partial \omega} \mathbf{H} \quad \text{(S76)}$$

is an integral over a volume $\mathcal{V}_1$ including the system inhomogeneity and I_2 is an integral of the same function over the region inside the perfectly matched layer (PML) in which the field decays due to the artificially absorbing medium of the PML. Here $\mathbf{H}$ is the corresponding magnetic field of the RS. We compare our normalization Eq. (S55) with Eq. (S75) by evaluating I_1, for which numerical values are provided in Ref. [15] for a TM mode of a gold sphere with radius $a = 0.1\,\mu$m having the wavelength $\lambda = 2\pi c/\omega = (0.607 + 0.239i)\,\mu$m. We use the dielectric constant of gold in the Drude model with the same parameters as in Ref. [15, 16]: $\epsilon(\omega) = 1 - \lambda^2/(0.15^2(1 + i0.075\lambda)$ with λ measured in μm.

The electric field of a TM mode has the form [4]

$$\mathbf{E}(\mathbf{r}) = \frac{A_l^{\mathrm{TM}}(k)}{\varepsilon(r) k r} \begin{pmatrix} l(l+1) R_l(r, k_n) Y_{lm}(\Omega) \\ \frac{\partial}{\partial r} r R_l(r,k) \frac{\partial}{\partial \theta} Y_{lm}(\Omega) \\ \frac{\partial}{\partial r} \frac{r R_l(r,k)}{\sin\theta} \frac{\partial}{\partial \varphi} Y_{lm}(\Omega) \end{pmatrix} \quad \text{(S77)}$$

in which $k = \omega/c$ is a solution of the secular equation for TM modes

$$\frac{1}{n_r} \frac{j_{l+1}(n_r ka)}{j_l(n_r ka)} = \frac{h_{l+1}(ka)}{h_l(ka)} + \frac{l+1}{ka} \left(1 - \frac{1}{n_r^2}\right), \quad \text{(S78)}$$

$R_l(r,k)$ is given by Eq. (S62) and $\varepsilon(r)$ by Eq. (S60) with $n_r^2 = \epsilon(\omega)$, taking any of the two roots for n_r. The normalization constant $A_l^{\mathrm{TM}}(k)$ calculated using the correct normalization Eq. (S55) has the form

$$\frac{n_r A_l^{\mathrm{TE}}}{\sqrt{2} A_l^{\mathrm{TM}}(k)} = \sqrt{\left[\frac{j_{l-1}(n_r ka)}{j_l(n_r ka)} - \frac{l}{n_r ka}\right]^2 + \frac{l(l+1)}{k^2 a^2} + \eta C_l(k)}, \quad \text{(S79)}$$

where A_l^{TE} is given by Eq. (S63), and the last term under the square root takes into account the effect of the dispersion, with

$$\eta = \frac{1}{\epsilon(\omega)} \frac{\partial(\omega^2 \epsilon(\omega))}{\partial(\omega^2)} - 1 \quad \text{(S80)}$$

and

$$(n_r^2 - 1) C_l(k) = \frac{2(l+1)}{k^2 a^2} + n_r^2 \left[\frac{j_{l+1}^2(n_r ka)}{j_l^2(n_r ka)} - \frac{j_{l+2}(n_r ka)}{j_l(n_r ka)}\right]. \quad \text{(S81)}$$

Note that the normalization constant $A_l^{\mathrm{TM}}(k)$ of a TM mode, defined by Eq. (S79), differs from from that for a dielectric sphere, given by Eq. (29) of Ref. [4], by the following two features: (i) it contains an extra factor of $\sqrt{2}$, here introduced in order to adapt it to the normalization Eqs. (S75), (S76) including both E- and H-field integrals, and (ii) as already noted, it takes into account the dispersion of the metal via the term $\eta C_l(k)$.

The corresponding magnetic field of the same RS has the form

$$i\mathbf{H}(\mathbf{r}) = A_l^{\mathrm{TM}}(k) R_l(r,k) \begin{pmatrix} 0 \\ \frac{1}{\sin\theta} \frac{\partial}{\partial \varphi} Y_{lm}(\Omega) \\ -\frac{\partial}{\partial \theta} Y_{lm}(\Omega) \end{pmatrix}. \quad \text{(S82)}$$

The integral $I_1 = I_{1E} + I_{1H}$ over a sphere of radius $R \geqslant a$ is then evaluated in the following way:

$$\begin{aligned} I_{1E} &= \int_{\mathcal{V}_R} d\mathbf{r}\, \mathbf{E} \cdot \frac{\partial(\omega \hat{\boldsymbol{\epsilon}}(\omega))}{\partial \omega} \mathbf{E} \\ &= \frac{\left[A_l^{\mathrm{TM}}(k)\right]^2}{k^2} \Bigg\{ [l(l+1)]^2 \int_0^R dr \frac{R_l^2(r,k) \beta(r)}{\varepsilon^2(r)} \int d\Omega Y_{lm}^2(\Omega) \\ &\quad + \int_0^R dr \frac{\beta(r)}{\varepsilon^2(r)} \left(\frac{\partial}{\partial r} r R_l(r,k)\right)^2 \\ &\quad \times \int d\Omega \left[\left(\frac{\partial Y_{lm}(\Omega)}{\partial \theta}\right)^2 + \frac{1}{\sin^2\theta} \left(\frac{\partial Y_{lm}(\Omega)}{\partial \varphi}\right)^2\right] \Bigg\} \\ &= \frac{\left[A_l^{\mathrm{TM}}(k)\right]^2}{k^2} l(l+1) \int_0^R dr \frac{\beta(r)}{\varepsilon^2(r)} \left[l(l+1) R_l^2 + (\partial_r r R_l)^2\right], \end{aligned} \quad \text{(S83)}$$

where

$$\beta(r) = \begin{cases} \dfrac{\partial(\omega \epsilon(\omega))}{\partial \omega} & \text{for } r \leqslant a \\ 1 & \text{for } r > a\,, \end{cases} \quad \text{(S84)}$$

and

$$I_{1H} = -\int_{\mathcal{V}_R} d\mathbf{r}\, \mathbf{H}\cdot\frac{\partial(\omega\hat{\boldsymbol{\mu}}(\omega))}{\partial\omega}\mathbf{H}$$
$$= \left[A_l^{\mathrm{TM}}(k)\right]^2\int_0^R r^2R_l^2dr\int d\Omega\left[(\partial_\theta Y_{lm})^2+\frac{(\partial_\varphi Y_{lm})^2}{\sin^2\theta}\right]$$
$$= \left[A_l^{\mathrm{TM}}(k)\right]^2 l(l+1)\int_0^R r^2R_l^2dr\,, \qquad \text{(S85)}$$

using $\hat{\boldsymbol{\mu}} = \hat{\mathbf{1}}$ everywhere. The integral in Eq. (S83) is calculated analytically using the Bessel equation and integration by parts:

$$\int dr\left[l(l+1)R_l^2+(\partial_r rR_l)^2\right]$$
$$= rR_l^2+r^2R_l\partial_rR_l+\varepsilon k^2\int r^2R_l^2dr\,, \qquad \text{(S86)}$$

where ε is constant in each area of space. The integral in the last term of Eqs. (S85) and (S86) is a known analytic integral:

$$\int x^2f_l^2(\alpha x)dx = \frac{x^3}{2}\left[f_l^2(\alpha x)-f_{l-1}(\alpha x)f_{l+1}(\alpha x)\right]\,, \qquad \text{(S87)}$$

in which $f_l(z)$ is any spherical Bessel function, $j_l(z)$ or $h_l(z)$, and α is a complex constant. We therefore find

$$I_{1E} = \frac{\left[A_l^{\mathrm{TM}}(k)\right]^2}{k^2}l(l+1)\left(\frac{1}{\epsilon(\omega)}\frac{\partial(\omega\epsilon(\omega))}{\partial\omega}I_{1E}^a+I_{1E}^R\right),$$
$$I_{1H} = \frac{\left[A_l^{\mathrm{TM}}(k)\right]^2}{k^2}l(l+1)\left(I_{1H}^a+I_{1H}^R\right), \qquad \text{(S88)}$$

where

$$I_{1E}^a = a(l+1)-\frac{n_rka^2}{2}(2l+3)\frac{j_{l+1}(n_rka)}{j_l(n_rka)}$$
$$+\frac{n_r^2k^2a^3}{2}\left(1+\frac{j_{l+1}^2(n_rka)}{j_l^2(n_rka)}\right),$$
$$I_{1E}^R = \frac{1}{2h_l^2(ka)}\left[-kr^2(2l+3)h_{l+1}(kr)h_l(kr)\right.$$
$$\left.+2r(l+1)h_l^2(kr)+k^2r^3\left(h_l^2(kr)+h_{l+1}^2(kr)\right)\right]_a^R,$$
$$I_{1H}^a = \left[\frac{k^2r^3}{2j_l^2(n_rka)}\left(j_l^2(n_rkr)-j_{l-1}(n_rkr)j_{l+1}(n_rkr)\right)\right]_0^a,$$
$$I_{1H}^R = \left[\frac{k^2r^3}{2h_l^2(ka)}\left(h_l^2(kr)-h_{l-1}(kr)h_{l+1}(kr)\right)\right]_a^R. \qquad \text{(S89)}$$

The values of $I_1 = I_{1E}+I_{1H}$ calculated using Eqs. (S88) and (S89) are shown in Table S1 and compared with the values I_1^{S} provided by Sauvan *et al.* [15, 16] for the same radii R of the sphere of integration. One can see an excellent agreement between the two approaches, with a relative error in the $10^{-7}-10^{-8}$ range. We note however that this result was obtained for a spherically symmetric system which is effectively one-dimensional (1D),

TABLE S1: The values of the integral Eq. (S76) calculated in the present work (I_1) and the relative difference between I_1 and the value I_1^{S} calculated by Sauvan *et al.* [15], for three integration radii R, for the mode with the wavelength $2\pi c/\omega = (0.607+0.239i)\,\mu$m in a gold nanosphere of radius $a = 0.1\,\mu$m.

R [μm]	I_1	I_1^{S}/I_1-1	$\lvert I_1^{\mathrm{S}}/I_1-1\rvert$
0.15	0.61936187690	$(5.66-1.29\,i)$	5.80×10^{-9}
	$-0.44899671324\,i$	$\times10^{-9}$	
1.0	6.56641919859	$(0.057+2.095\,i)$	2.095×10^{-8}
	$+0.49127433385\,i$	$\times10^{-8}$	
2.0	1052.29778832465	$(0.100+4.468\,i)$	4.469×10^{-8}
	$-1235.22683098918\,i$	$\times10^{-8}$	

for which the calculation in Ref. [15] was done analytically [16] that actually explains the excellent agreement with the strict result. In a full 3D calculation the use of a PML may lead to more significant errors. For instance, using the approach of Ref. 15 a deviation of about 2% of the PF from the direct numerical evaluation of the GF was found [15, 17] for an optical mode in a gold rod with cylindrical symmetry. A more detailed comparison of this numerical normalization method with the exact normalization would be interesting.

V. DETAILS OF THE PURCELL FACTOR CALCULATION

In this section we provide some details of our calculation of the PF for a dielectric spherical resonator in vacuum, of radius a and refractive index n_r; numerical results are presented here and in the main text. The PF is expressed in terms of the mode volumes via Eq. (9), and the mode volume of a RS is given by Eq. (2) of the main text, in terms of its normalized electric field. The latter has an explicit analytic form for a spherical resonator, which is given by Eq. (S61) for TE and by Eq. (S77) for TM polarization. The normalization constants are given by Eqs. (S63) and (S79), where in the latter we remove the factor $\sqrt{2}$ which was introduced for the comparison with Ref. 15. Static modes do not contribute to the PF as noted in Sec. I and thus are not considered here.

Owing to the spherical symmetry of the resonator, RS eigenfrequencies are $2l+1$ degenerate with respect to the azimuthal quantum number m (here l is the orbital quantum number). Therefore, for each set of degenerate RSs, we introduce a collective mode volume V_l defined as

$$\frac{\mu^2}{V_l} = \sum_{m=-l}^{l}\left[\boldsymbol{\mu}\cdot\mathbf{E}_{lm}(\mathbf{r})\right]^2\,, \qquad \text{(S90)}$$

where the quantum numbers l and m are shown explicitly but the RS index n is dropped for brevity of notation. Then, using the vector components of the dipole moment

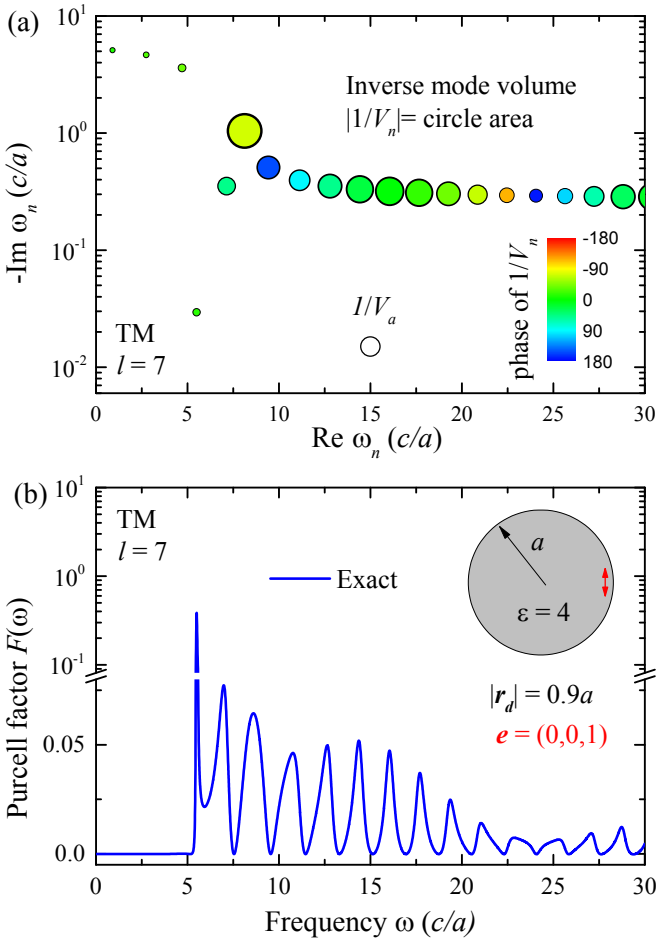

FIG. S3: (a) Mode volumes for a dielectric sphere in vacuum, with permittivity $\varepsilon = 4$ and radius a, for $l = 7$ TM modes, for a point dipole placed at $|\mathbf{r}_d| = 0.9a$ with direction $\mathbf{e} = (0, 0, 1)$, in spherical coordinates (see sketch). The mode volume is presented as the sum of the inverse mode volume over all degenerate states $m = -l, \ldots, l$. Its amplitude is shown by the circle area and its phase by the color. The volume of the sphere $V_a = 4\pi a^3/3$ is shown for comparison. The position of the circles in the complex frequency plane is given by the mode eigenfrequency ω_n. (b) Partial Purcell factor as a function of the dipole frequency ω, calculated for the geometry of (a).

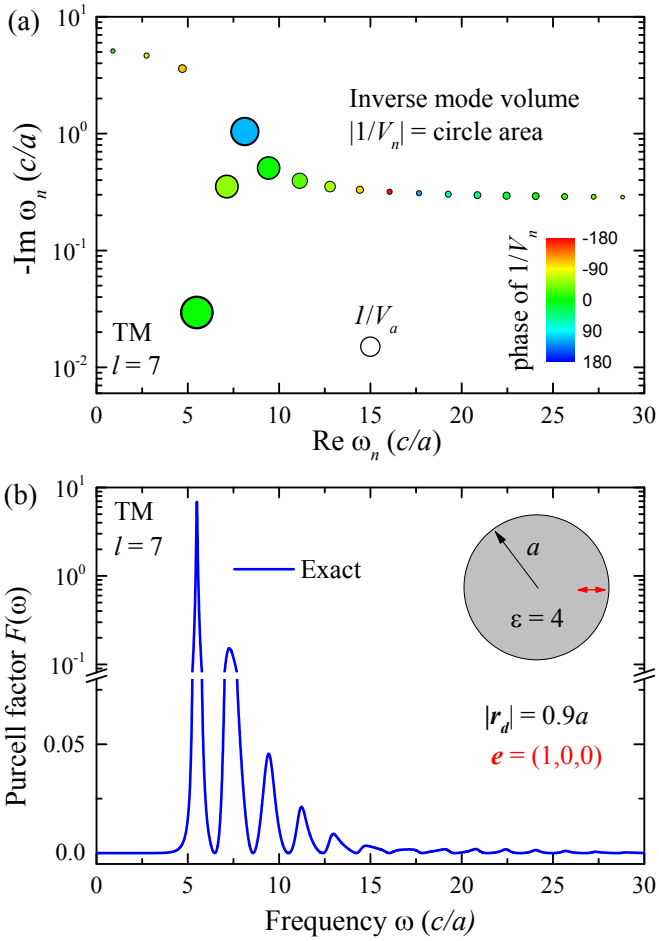

FIG. S4: As in Fig. S3 but for an orthogonal direction of the dipole: $\mathbf{e} = (1, 0, 0)$.

in spherical coordinates,

$$\boldsymbol{\mu} = \mu_r \mathbf{e}_r + \mu_\theta \mathbf{e}_\theta + \mu_\varphi \mathbf{e}_\varphi \,, \tag{S91}$$

and the sum rules for spherical harmonics, we obtain

$$\begin{aligned}\frac{\mu^2}{V_l^{\mathrm{TE}}} &= \frac{\left[E_l^{\mathrm{TE}}(r)\right]^2}{l(l+1)} \sum_{m=-l}^{l} \left[\mu_\theta^2 \left(\frac{1}{\sin\theta}\frac{\partial Y_{lm}}{\partial \varphi}\right)^2 + \mu_\varphi^2 \left(\frac{\partial Y_{lm}}{\partial \theta}\right)^2\right] \\ &= \left[E_l^{\mathrm{TE}}(r)\right]^2 \frac{2l+1}{8\pi}(\mu_\theta^2 + \mu_\varphi^2)\end{aligned} \tag{S92}$$

for TE modes, and

$$\begin{aligned}\frac{\mu^2}{V_l^{\mathrm{TM}}} &= \left[E_l^{\mathrm{TM1}}(r)\right]^2 l(l+1) \sum_{m=-l}^{l} \mu_r^2 Y_{lm}^2 \\ &\quad + \frac{\left[E_l^{\mathrm{TM2}}(r)\right]^2}{l(l+1)} \sum_{m=-l}^{l} \left[\mu_\theta^2 \left(\frac{\partial Y_{lm}}{\partial \theta}\right)^2 + \mu_\varphi^2 \left(\frac{1}{\sin\theta}\frac{\partial Y_{lm}}{\partial \varphi}\right)^2\right] \\ &= \left[E_l^{\mathrm{TM1}}(r)\right]^2 l(l+1)\frac{2l+1}{4\pi}\mu_r^2 \\ &\quad + \left[E_l^{\mathrm{TM2}}(r)\right]^2 \frac{2l+1}{8\pi}(\mu_\theta^2 + \mu_\varphi^2)\end{aligned} \tag{S93}$$

for TM modes, where

$$\begin{aligned}E_l^{\mathrm{TE}}(r) &= \sqrt{\frac{2}{(n_r^2-1)a^3}} R_l(r)\,, \\ E_l^{\mathrm{TM1}}(r) &= \sqrt{\frac{2}{(n_r^2-1)a^3 D_l}} \frac{1}{n_r kr} R_l(r)\,, \\ E_l^{\mathrm{TM2}}(r) &= \sqrt{\frac{2}{(n_r^2-1)a^3 D_l}} \frac{1}{n_r kr} \frac{\partial}{\partial r} r R_l(r)\,,\end{aligned} \tag{S94}$$

$R_l(r) = j_l(n_r kr)/j_l(n_r ka)$, r is the position of the dipole (inside the dielectric sphere), and

$$D_l = \left[\frac{j_{l-1}(n_r ka)}{j_l(n_r ka)} - \frac{l}{n_r ka}\right]^2 + \frac{l(l+1)}{k^2 a^2}\,. \qquad \text{(S95)}$$

The collective mode volumes of several RSs with $l = 7$, calculated using Eqs. (S92)–(S94), are shown in Fig. 1(a) of the main text for TE polarization and in Figs. S3(a) and S4(a) for TM polarization and two different directions of the dipole. We note that the fundamental $n = 1$ WGMs in TE and TM polarizations, which have quite similar Q-factors of the order of 100, have very different mode volumes for a given direction of the dipole. Indeed, for an azimuthal dipole direction $\mathbf{e} = (0, 0, 1)$ the effective volume of the TE mode is much smaller than the one of the TM mode. This is because the electric field in TM polarization is mostly in radial direction, with only a small azimuthal component. For a radial direction of the dipole $\mathbf{e} = (1, 0, 0)$ instead, the TM mode has a much smaller mode volume, comparable to that of the TE mode for $\mathbf{e} = (0, 0, 1)$, as seen by comparing Fig. 1(a) and Fig. S4(a). The partial PFs due to all $l = 7$ modes within the spectral range up to $\omega_n a/c \sim 40$ are shown separately, in Fig. 1(b) for TE and in Figs. S3(b) and S4(b) for TM polarization. These figures demonstrate the strong dependence of the PF on the dipole orientation, as discussed above. Summing over all different l components and averaging over all possible directions of the dipole, we obtain the full PF for this system which is demonstrated in Fig. 2 of the main text.

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